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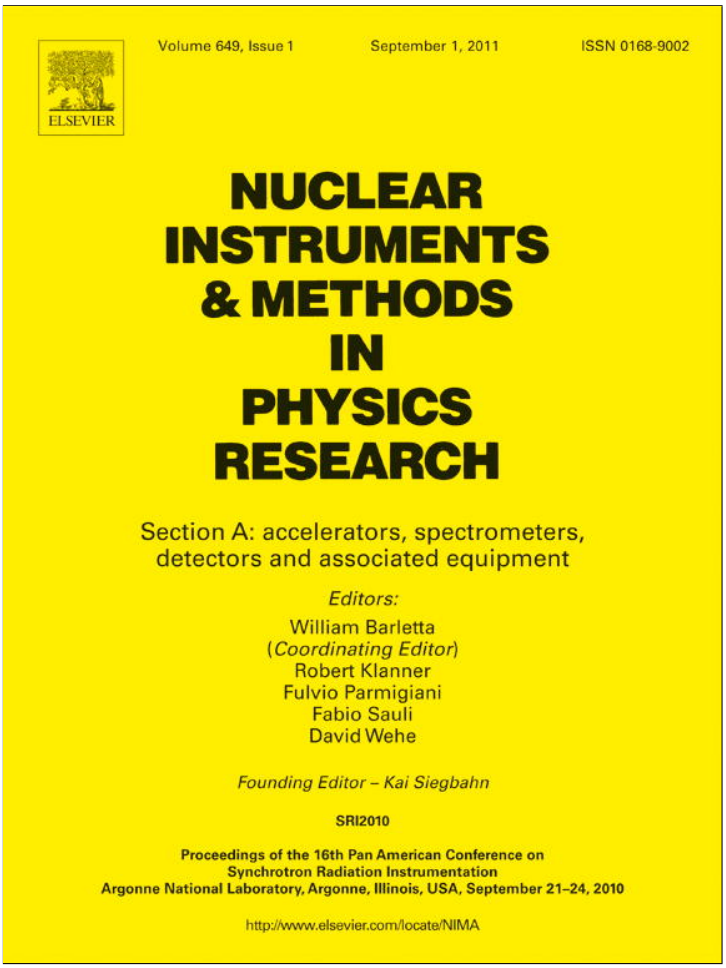

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A technique for high-frequency laser-pump X-ray probe experiments at the APS

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ABSTRACT

When a short-pulse laser beam is absorbed in a crystal, the heat or large electric field can induce time-dependent strain waves which propagate in the material at the speed of sound. At a synchrotron, the repetition rate of the X-ray source (MHz) and the laser (kHz) is often mismatched by several orders of magnitude leading to a very inefficient use of the X-ray probe beam. In this paper, we will show how one can synchronize a femtosecond 88 MHz Ti:Sapphire laser to the APS running at the same repetition rate in 324-bunch mode. This efficient use of the X-rays enabled us to measure coherent diffraction patterns from nanoparticle of ZnO as a function of the Bragg angle and time delay between the laser-pump and X-ray probe beams. Significant time-dependent strain can be created with a few nanoJoule per pulse when the nanoparticle is centered in a tight laser focus of a few microns.

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1. Introduction

The interaction of light with nanomaterials is rich in novel effects that come from the interaction between the surface and bulk of the objects, as well as from quantum confinement effects. In semiconducting nanocrystals, the light absorption can be tuned by the size of the particle. The interaction of light in these crystals can yield very large strain fields of several percent which may be resolved by hard X-ray or electron diffraction [1,2]. These preliminary works illuminate an ensemble of nanowire and measure their average time-resolved response.

Time-resolved diffraction from bulk single crystal and thin films following a kHz pulsed laser excitation is a well-developed technique at synchrotrons providing time-resolution as short as the bunch duration of 100 ps (see Ref. [3] and references within). In this paper, we show how one may increase the sensitivity of these laser-pump X-ray probe experiments [1–3] to resolve the response of isolated nanocrystals. At the Advanced Photon Source (APS), the bunch fill pattern for 6 weeks of the year is a 50 ps-long bunch with a repetition rate of 88 MHz. A Commercial Ti:Sapphire oscillator is routinely synchronized to this RF range at synchrotrons [3]. This operation mode of the APS allows one to use all the time-averaged brightness and flux of the synchrotron in time-resolved stroboscopic laser-pump and X-ray probe experiments. This paper describes the instrumentation required to measure time-resolved coherent microdiffraction from ZnO crystals.

2. Experimental

The experiments were performed at beamline 7ID of the APS [3]. Preliminary experiments characterizing the sample were performed at beamline 34ID of the APS [4,5]. A monochromatic 9 keV X-ray beam with diamond (1 1 1) energy resolution is focused on the sample by a Kirkpatrick–Baez mirror system to a focal spot of about 15 μm × 15 μm. The incident beam on this KB system is limited by slits set to 200 μm × 50 μm in the vertical and horizontal direction, respectively. The vertical dimension matched the transverse coherence length of the 7ID beam at 9 keV, while the horizontal slit opening was about five transverse coherence lengths. The coherent flux for this experiment was about 5×10^{11} ph/s at a ring current of 100 mA, but the typical micron-cubed sample only intercepted a fraction of one percent of this coherent flux. The sample size is much smaller than the coherence length of the focused beam and thus is illuminated coherently. The diamond (1 1 1) monochromator provides about 0.5 eV energy resolution thus its longitudinal coherence length is about 1.2 μm [5].

A femtosecond Coherent Micra Ti:Sapphire oscillator is phased-locked to the APS Radio Frequency (RF) pulsing at 88 MHz. The 352 MHz RF is distributed to the beamline from

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the master clock of the APS through a single-mode fiber. A Coherent Synchrolock AP system stabilizes the jitter of the laser and a RF subharmonics signal (RF frequency divided by four) to 250 fs RMS. The laser can be delayed from the RF by a programmable delay line (Colby Instruments PDL-100A-20NS) in steps of 0.5 ps over a range of 20 ns.

The 20-fs-long-laser is focused on the sample with commercial long-working-distance objectives with X5 magnification [6]. We could focus up to 290 mW of 800 nm light into a 6 μm spot size leading to a peak fluence of 11.7 mJ/cm^2 [6]. This peak intensity is comparable to recent time-resolved fs electron diffraction results on ZnO nanocrystals [1].

The ZnO microcrystals growth was described elsewhere [4]. They are grown by a vapor flow chemical vapor deposition onto Si substrates, then annealed in an oxygen-rich atmosphere. The microcrystals are typically hexagonally faceted nanorods with a length of 1–2 μm and a diameter of 500 nm. They were coated with a thin Ni film to absorb the infrared light. They are randomly oriented on the Si substrate with a typical *c*-axis orientation along the surface normal of the substrate.

The sample were mounted on the sample stage of a 6-circle Huber diffractometer. A Huber XYZ translation stage mounted on the phi circle of the goniometer enables one to center crystals in the laser and X-ray beams. A thermoelectrically cooled Princeton Instruments PI-LCX:1300 CCD camera with a deep depletion EEV chip is used to measure Coherent X-ray patterns from the sample. This camera is ungated, and directly detects X-rays in the Si depletion layer. It is shuttered by a fast millisecond shutter (NM Laser Products, model LS500FNC). Because the laser and X-rays share the same frequency, any standard synchrotron detector can be used. This is an improvement in quantum efficiency over previous work done with optically coupled CCD cameras observing the visible light from X-ray sensitive single crystal Ce doped YAG [6]. The sample to CCD distance is 1.7 m, and its pixel size is 20 μm. The phi circle oriented with a vertical axis of rotation was used to diffract X-rays in a horizontal plane. Since the 2θ angle is around 25°, the polarization losses are only about 20%.

The coherent diffraction pattern from a single crystal is aligned by first capturing the powder diffraction pattern of many nanocrystals with the CCD detector, then by isolating one crystal by moving the sample in the beam such that a symmetric diffraction pattern is observed on the CCD. The rocking curve is then peaked up by scanning the crystal Bragg angle.

A key challenge is to overlap the laser and X-rays both in time and space. Adjusting the time delay between the laser and X-rays was accomplished by measuring separately the time difference between the laser and X-rays on an InGaAs photodiode centered in the sample position. We triggered a 16 GHz Tektronix DPO 71604 digital oscilloscope with a laser fiducial signal generated by a fraction of the light transmitted through a thin laser pellicle (Thorlabs BP133 with 33% transmission at 633 nm) absorbed on another InGaAs diode. An identical diode is used at the sample position, but since the response to X-rays is weaker than it is for light, the diode signal is amplified by an 18 GHz amplifier with a gain of 26 dB (Mini-Circuits ZVA-183-S+). The glass cover of the diode assembly has been removed to improve the diode response to X-rays. It is then straightforward to measure the X-ray and laser signals at the sample position and adjust the time delay between the laser and X-ray to a few picosecond RMS. The diode sensitive area is a crystal of about 40 μm, so this time-resolved signal amplitude can be optimized to overlap the X-ray and laser beams spatially. This provides a coarse adjustment. We have found that the best way to center the nanoparticle in the X-ray and laser beams is to optimize the time-average strain induced by laser heating. The laser heating displaces the diffracted beam on the CCD by several pixels so this displacement is optimized when the overlap is ideal.

3. Results and discussion

Fig. 1 shows the laser trigger diode signal and the sample position diode signal with laser and X-ray signals offset by 2.0 ns. The reference signal has a period of 11.3 ns as expected. The laser on the sample was attenuated by neutral density filters to enable direct comparison between the X-ray and laser signals. The X-ray signal amplitude here is around −6.5 mV, and was delayed by the Colby programmable delay line. The jitter between the X-ray and laser rise time is a few picoseconds RMS as measured with the oscilloscope.

Fig. 2 shows the effect of turning the laser on or off onto the ZnO (0 0 2) Bragg speckle pattern at time zero. The X-ray energy was set to 9.000 keV. The peak of the speckle pattern is displaced by 6.5 pixels, which corresponds to a change in 2θ angle of 6.5∗20 μm/1.7 m = 76.5 μrad. Taking a derivative of Bragg's law, one can show that the strain is $\Delta a/a = \alpha\Delta T = \Delta\theta/\tan\theta$, where a is the equilibrium lattice constant, α is the coefficient of thermal expansion, and ΔT is the change in temperature. Using the value of α along the *c*-axis for ZnO (2.92×10^{-6}/K), and the equilibrium Bragg angle, one finds the crystal heated up by 47 K. The fringe visibility of nearly 90% shows that the setup provides adequate coherence to observe coherent diffraction patterns. Subtle changes in the speckle pattern are notable, and may be a sign of strain fields caused by the laser heating. A detailed quantitative

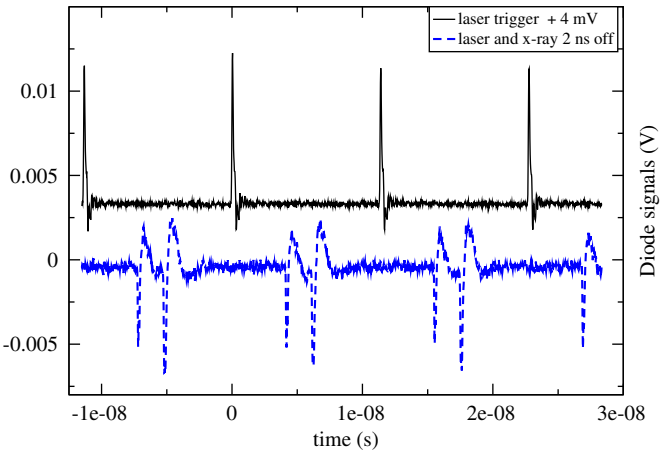

Fig. 1. Laser reference trigger signal (solid line) offset by 4 mV, and the X-ray and laser signal delayed by 2 ns at the sample position (dashed line).

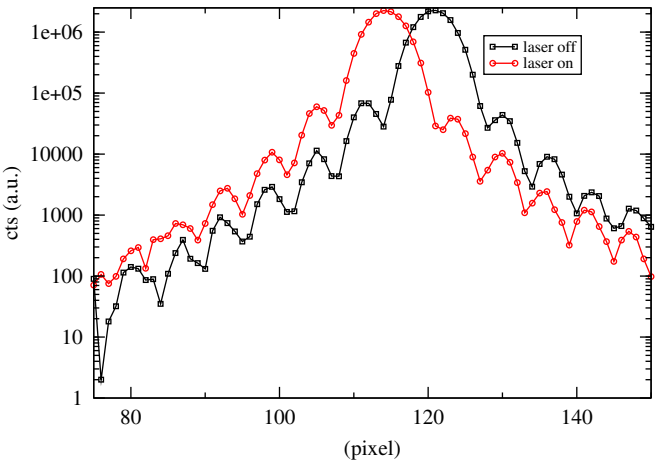

Fig. 2. Slices of two speckle patterns with laser on and laser off.

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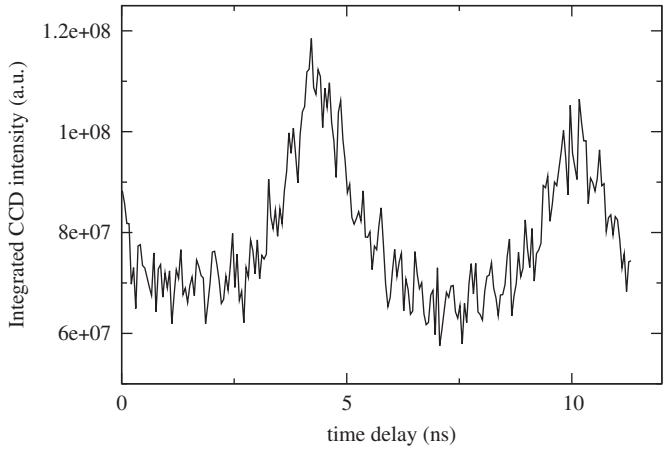

Fig. 3. Time-resolved integrated intensity oscillations.

analysis of this effect is beyond the scope of this paper, but the result shows how one can optimize the laser and X-ray alignment as stated in the previous section.

Fig. 3 shows the integrated intensity from a prealigned ZnO nanocrystal as a function of the delay time between the laser and X-rays. The integrated intensity on the CCD shows 50% variations as a function of time with a period of 5.7 ns. These oscillations may be mechanical vibrations of the crystal as observed by Mariager et al. [2]. More detailed studies are required to understand the dynamics.

We have shown how one can measure time-resolved coherent diffraction patterns from isolated ZnO nanocrystals when the APS is running in 324-bunch mode, using an 88 MHz oscillator and stroboscopic scanning of the time delay. With a novel laser source such as a Ti:Sapphire long-cavity oscillator, one could get pulse energies 50 times higher than this study so that one could operate in the standard 24-bunch mode of the APS. This would allow for longer time delays to be probed, as well as allow more time for the sample to cool.

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